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Three-Tier Hierarchical Structures for Extreme Pool Boiling Heat Transfer Performance

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Peer reviewed

- 1 Three-Tier Hierarchical Structures for Extreme Pool Boiling Heat Transfer
- 2 **Performance** 3

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13 Keywords: Hierarchical structure, Microstructure, Nanostructure, Critical heat flux, Heat

- 14 transfer coefficient, Phase change heat transfer
- 15

16 Boiling is an effective energy transfer process with substantial utility in energy applications. 17 Boiling performance is described mainly by the heat transfer coefficient (HTC) and critical 18 heat flux (CHF). Recent efforts for the simultaneous enhancement of HTC and CHF have 19 been limited by an intrinsic trade-off between them — HTC enhancement requires high 20 nucleation site density, which can increase bubble coalescence resulting in limited CHF 21 enhancement. In this work, we overcome this trade-off by designing three-tier hierarchical 22 structures. We minimize bubble coalescence to enhance CHF by defining nucleation sites with 23 microcavities interspersed within hemi-wicking structures. Meanwhile, the reduced nucleation 24 site density is compensated for by incorporating nanostructures that promote evaporation for HTC enhancement. Our hierarchical structures demonstrate the simultaneous enhancement of 25 26 HTC and CHF up to 389% and 138%, respectively, compared to a smooth surface. This 27 extreme boiling performance can lead to significant energy savings in a variety of boiling 28 applications.

29

The global demand for electrical power has been continuously growing ~3% annually during the past decade.<sup>[1]</sup> Over 90% of this electrical power is generated by steam turbines,<sup>[2]</sup> where boiling generates the steam. Steam generation is also essential in sustainability such as for

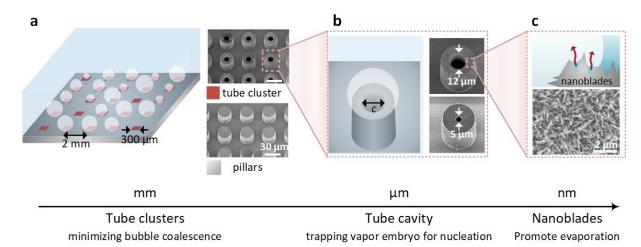
food and chemical processing, water purification, sterilization, and harvesting.<sup>[3]</sup> Furthermore,
the high heat dissipation capability of boiling enables such processes for thermal management
of high flux energy applications such as concentrated photovoltaics, fast-charging batteries,
data centers, and integrated electronics.<sup>[4]</sup> The continuing growth in energy demands puts an
increased emphasis on achieving even larger enhancements in boiling performance.

6 The heat transfer coefficient (HTC, h) and critical heat flux (CHF, q } rsub {CHF  $\dot{i}$ ) are two 7 major parameters that quantify boiling performance. The HTC describes the efficiency of boiling heat transfer, defined as the ratio of heat flux (q) to the wall superheat  $(\Delta T_w)$ , i.e., 8 9 h=q / { $\Delta$  {T} rsub {w}  $\dot{\iota}$ . Here  $\Delta T_w$  is the temperature difference between the boiling 10 surface and the saturated liquid. In the nucleate boiling regime, the heat flux increases with 11 the wall superheat. However, when the heat flux is sufficiently high, excessive vapor bubbles 12 nucleated on the boiling surface prevent the liquid from rewetting the surface and, in turn, 13 form an insulating vapor film over the surface. This vapor film becomes a thermal barrier that 14 leads to a drastic increase in wall superheat and burnout of a boiling system. This transition 15 from nucleate boiling to film boiling is known as the boiling crisis, where the maximum heat 16 flux is CHF. Enhancing CHF, therefore, can either enable larger safety margins or extend the 17 operational heat flux range for boiling systems.<sup>[5]</sup>

Recent efforts to enhance boiling heat transfer have focused on engineering the working fluid 18 19 or surface properties.<sup>[6]</sup> In particular, engineering surface structures has received greater 20 attention owing to the constraints on chemical compatibility or operational conditions which 21 can limit the choice of the working fluid. Representative examples of surface structures that 22 effectively enhance CHF are known to be hemi-wicking surfaces such as micropillars and 23 nanowires.<sup>[7]</sup> These structures enhance CHF by harnessing thin-film evaporation around pillars and capillary-fed wicking through the structures.<sup>[8]</sup> Surfaces with microcavities, on the 24 25 other hand, have shown improved HTC by trapping vapor embryos that promote nucleation.<sup>[9]</sup>

1 Recently, a combination of microtube and micropillar structures, referred to as tube-clusters 2 in pillars (TIP), has shown the ability to tune the HTC and CHF by controlling bubble coalescence while maintaining capillary wicking.<sup>[10]</sup> Despite the controllability, achieving 3 extreme enhancement of HTC and CHF simultaneously remains challenging due to the 4 intrinsic trade-off between HTC and CHF associated with nucleation site density. For 5 example, high nucleation site density may increase HTC but decrease CHF because extensive 6 7 bubble coalescence hinders the capillary wicking performance, while the reduced number of 8 nucleation sites will limit the HTC enhancement. In addition, certain copper-based engineered surfaces have shown simultaneous enhancements of HTC and CHF,<sup>[11]</sup> but the change in 9 surface morphology and chemical composition of copper surfaces by in situ oxidation during 10 boiling has made the enhancement mechanism unclear.<sup>[12]</sup> 11

12 In this work, we overcome this trade-off and demonstrate an approach to achieve 13 simultaneous extreme enhancement of HTC and CHF based on chemically stable SiO<sub>2</sub> 14 surfaces. By designing a three-tier hierarchically structured TIP surface (h-TIP), we can 15 control vapor nucleation at multiple length scales while providing capillary wicking (Figure 1). For CHF enhancement, we minimized bubble coalescence and exploited capillary wicking 16 17 by defining separated nucleation sites with tube clusters (shaded dark-red area in Figure 1a). 18 For HTC enhancement, on the other hand, we promoted vapor nucleation with microscale 19 cavities (Figure 1b), where evaporation could be further enhanced by extending the liquid-20 vapor interfaces on nanostructures (Figure 1c). Our strategy to enhance HTC does not have 21 the detrimental effect of continuing to increase the nucleation site density and as a result, 22 prevents the liquid rewetting process.



1

2 Figure 1. Schematics and scanning electron microscopy (SEM) images of a hierarchically 3 structured TIP surface (h-TIP) that exhibits capillary wicking while controlling vapor 4 nucleation using multiple length scales. (a) The h-TIP surface consists of hierarchical tube 5 clusters interspersed in hierarchical pillar arrays. Left panel: 2 mm pitch between the tube 6 clusters separates bubbles and minimizes bubble coalescence. The dark-red patterned regions represent the hierarchical tube clusters whereas the grey region is covered by hierarchical 7 8 pillar structures. Right panel: SEM images of the hierarchical tube structures (top) and 9 hierarchical pillar structures (bottom). Scale bars are 30 µm. (b) Microscale cavity at the 10 center of tube structure traps vapor embryo and promotes nucleation. Left panel: schematic of 11 bubble nucleation on the top of a tube structure. Right panel: SEM images of a hierarchical 12 tube with a cavity diameter of 12 µm (top) and 5 µm (bottom). (c) Nanoblades on top of microstructures augment liquid evaporation under the bubbles. Top panel: schematic of the 13 14 extended liquid-vapor interface on the nanostructures. Bottom panel: SEM image of the 15 nanostructures created all over the h-TIP and h-Tube surfaces including their tubes, pillars, 16 and flat base.

17

18 We fabricated hierarchically structured TIP surfaces (h-TIP) along with uniform arrays of 19 hierarchically structured tubes (h-Tube), which consist of only tubes, to investigate the

1 effectiveness of bubble separation by TIP structures in the presence of nanostructures. First, 2 microscale structures were created through photolithography and deep-reactive ion etching processes on silicon wafers. The outer diameter, height, and pitch of microtubes and 3 micropillars were fixed at 22, 30, and 40 µm, respectively. To investigate the effects of cavity 4 size on the onset of nucleate boiling temperature, two cavity diameters of 5 and 12 µm were 5 fabricated (SEM images in Figure 1b) for both h-Tube and h-TIP surfaces. According to the 6 7 theoretical analysis for active cavity sizes for nucleate boiling,<sup>[13]</sup> 5 and 12 µm cavities were chosen to initiate vapor nucleation at 11 °C and 5 °C wall superheat, respectively.<sup>[10]</sup> The 8 9 cluster-to-cluster pitch was set to 2 mm based on the capillary length of water ( $\approx 2.5$  mm) 10 (Figure 1a), which has been found as an optimal distance between nucleation sites for effective separation of vapor bubbles in previous works.<sup>[10, 14]</sup> On top of the microstructured 11 12 surfaces, sharp blade-like cupric oxide (CuO) nanostructures were created by sputtering a 500 13 nm copper layer over the microstructures followed by an oxidation in an alkali solution (NaClO<sub>2</sub>, NaOH, Na<sub>3</sub>PO<sub>4</sub>, and deionized water with 3.75:5:10:100 wt.%) at 95 °C for 2 14 15 minutes (SEM image in Figure 1c).<sup>[15]</sup> The structural durability of CuO nanostructures during boiling have been demonstrated in previous studies.<sup>[16]</sup> At the end, we deposited a 20 nm 16 silicon dioxide (SiO<sub>2</sub>) layer using atomic layer deposition to ensure uniform hydrophilicity 17 along the entire hierarchical structure. 18

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The pool boiling experiments were performed with saturated high-purity deionized water at atmospheric conditions by applying heat flux through a serpentine platinum (Pt) heater on the backside of each sample. This Pt heater was also used for thermometry based on its linear correlation between electrical resistance and temperature. Prior to boiling, all surfaces were cleaned with solvents followed by an argon plasma to remove organic contaminants.<sup>[17]</sup> The

1 bubble dynamics during boiling was captured by a high-speed camera (Phantom v7.1, Vision

- 2 Research) with up to 4000 frames per second.
- 3

Figure 2a shows pool boiling curves, i.e., heat flux as a function of wall superheat, of h-TIP 4 and h-Tube surfaces along with microstructured surfaces with the same microscale 5 dimensions such as micropillars, microtubes, and TIP surfaces from the previous work for 6 comparison.<sup>[10]</sup> Each surface is named with its structure and the cavity diameter; for example, 7 h-TIP 12 µm is a h-TIP surface that has 12 µm diameter cavity in each tube clusters. We also 8 plotted HTC as a function of heat flux in Figure 2b. All hierarchical structures showed 9 10 enhanced HTC and CHF values compared with their microstructure counterparts without 11 nanostructures. For example, h-TIP surfaces significantly enhanced HTC values while 12 maintaining the high CHF of TIP surfaces. Compared with up to 209% enhancement (relative to a flat surface) of maximum HTC ( $h_{max}$ ) on normal TIP surfaces, h-TIP 5 µm and h-TIP 12 13 µm achieved 289 and 389% enhancement, respectively. At the same time, CHF values of h-14 15 TIP 5 µm and h-TIP 12 µm were enhanced 138 and 125%, respectively. In fact, the boiling 16 curves of h-TIP surfaces shifted left of the boiling curves of normal TIP surfaces. This simultaneous enhancement of HTC and CHF on h-TIP structures was achieved because the 17 nanostructures extended the liquid-vapor interface. As a result, this effect promoted 18 19 evaporation and enhanced HTC. Meanwhile, the nucleation site density was maintained 20 (Figure 2c and 2d) to prevent hindering of the liquid rewetting, which would limit CHF 21 enhancement. We attribute two mechanisms within the nanostructures for promoting 22 evaporation and bubble departure frequency. First, the liquid imbibed in the nanostructures can evaporate quickly under the vapor bubbles because of the nanoscale liquid film thickness 23 24 (mechanism (1) in Figure 2d). Second, when the nanostructures are filled with vapor, the

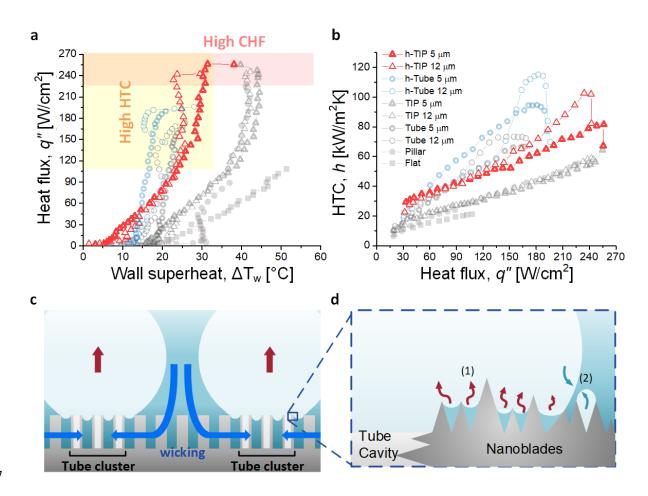
vapor can diffuse and merge with a growing bubble above them, which will accelerate the
 bubble growth and departure frequency (mechanism (2) in Figure 2d).

3 A similar enhancement was observed on h-Tube surfaces compared with normal microtube surfaces due to the enhanced evaporation from the nanostructures. The maximum HTC value 4 of h-Tube 5 µm and h-Tube 12 µm, for example, were 351 and 448% enhancement, 5 respectively. This result is significant improvement compared with the 244% enhancements of 6  $h_{max}$  on normal microtube arrays. In addition to the HTC enhancement, h-Tube surfaces 7 8 demonstrated higher CHF enhancement than microtube surfaces, where h-Tube 5 µm and h-9 Tube 12 µm achieved 78 and 82% CHF enhancement, respectively, while Tube 12 µm 10 showed 62% CHF enhancement. Because the extensively high nucleation site density 11 primarily triggered the boiling crises on h-Tube and microtube surfaces, the addition of 12 nanostructures was able to enhance CHF as well as HTC by providing additional evaporation 13 heat flux for a similar nucleation site density near CHF. Yet, the CHF enhancement was still limited compared to TIP and h-TIP surfaces. The pool boiling results of h-TIP and h-Tube 14 15 surfaces confirm that our h-TIP surfaces maintained the high CHF values of TIP surfaces 16 while significantly improving the HTC because of the effective separation of liquid and vapor 17 paths during boiling, which also allows surfaces to exploit the capillary wicking completely 18 (Figure 2c).

19 Another interesting observation to note was that hierarchical surfaces with 12  $\mu$ m cavity 20 diameters (h-Tube 12  $\mu$ m and h-TIP 12  $\mu$ m) showed a decrease in wall superheat near 20 °C 21 while the heat flux increased, a phenomenon known as boiling inversion. However, a similar 22 phenomenon was not observed on hierarchical surfaces with the 5  $\mu$ m cavity diameter. While 23 previous works attributed the boiling inversion primarily to enhanced macro-convection 24 driven by effective separation of liquid-vapor paths,<sup>[18]</sup> another work showed that the boiling 25 inversion disappeared when boiling curves were obtained by decreasing heat flux,<sup>[19]</sup> which

cannot be explained by the macro-convection hypothesis. Interestingly, our h-TIP 12 μm
 surface showed boiling inversion even on the boiling curve obtained by decreasing heat flux.
 Understanding the boiling inversion mechanism is still limited and needs further
 investigation. A detailed discussion on experimental repeatability and boiling inversion is
 available in Section II in the Supporting Information.







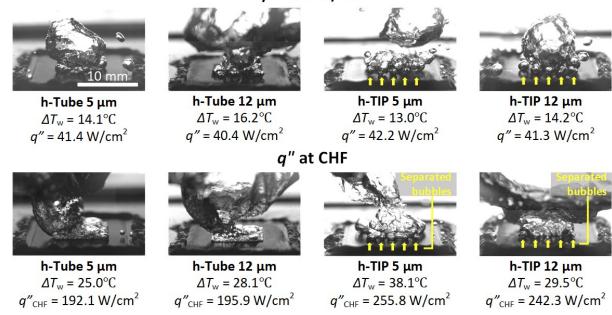
**Figure 2.** Pool boiling results and schematics of boiling heat transfer enhancement on h-TIP surfaces. (a) Pool boiling curves of saturated water on h-Tube and h-TIP surfaces compared with microstructured surfaces without nanostructures. (b) HTC as a function of heat flux. The experimental uncertainty is smaller than the marker size. (c) Schematic of the separated liquid-vapor paths during boiling on an h-TIP surface. Bubbles grow on top of and depart from tube clusters, while liquids coming outside tube clusters rewet the surface by capillary

wicking. (d) Schematic explaining the two mechanisms for the nanostructure-induced
 evaporation: (1) rapid evaporation of thin liquid films due to their nanoscale thickness and (2)
 diffusion of vapor trapped in nanostructures, which accelerates bubble growth and departure.

4

5 We confirmed the different behavior of bubble interactions on h-Tube and h-TIP surfaces by capturing the bubble dynamics with a high-speed camera (Phantom v7.1, Vision Research) up 6 7 to 4000 frames per second. High-speed movies are available in the Supporting Information. 8 Figure 3 shows images capturing the bubble dynamics on h-Tube and h-TIP surfaces at a 9 moderate heat flux of  $\approx 40$  W cm<sup>-2</sup> (the first row of Figure 3) and at CHF (the second row of 10 Figure 3). Since microscale cavities promote nucleation, significantly higher nucleation site 11 densities were observed on h-Tube surfaces compared to h-TIP surfaces. Accordingly, 12 bubbles on h-Tube surfaces showed intensive interactions with each other. In the case of h-TIP surfaces, on the contrary, bubbles nucleated from spatially separated tube clusters, which 13 led to a reduced bubble coalescence. This behavior continued until CHF was reached. We 14 observed that vapor films formed over h-Tube surfaces at CHF, which suggests that CHF 15 16 enhancement was limited because of the high nucleation site density. The separation of 17 nucleating bubbles on h-TIP surfaces, on the other hand, was effective even at CHF; therefore, 18 the CHF on h-TIP surfaces was enhanced until it was limited by capillary wicking. This 19 observation supports our boiling curve analysis that h-TIP surfaces enhanced HTC without 20 increasing the nucleation site density, resulting in simultaneous enhancement of HTC and 21 CHF.





**Figure 3.** Images of bubbles on h-Tube and h-TIP surfaces during boiling captured by a highspeed camera with a 19° tilt angle and up to 4000 frames per second. First and second rows show bubble dynamics at a heat flux of  $\approx 40$  W cm<sup>-2</sup> and at CHF, respectively. h-TIP surfaces showed effectively separated bubbles even at CHF (highlighted with yellow arrows), while h-Tube surfaces showed extensive bubble coalescence which led to the formation of vapor films at CHF. High-speed movies are available in the Supporting Information.

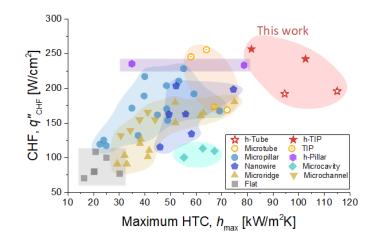
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9 In Figure 4, we compared CHF (q ] rsub {CHF  $\dot{i}$ ) and the maximum HTC ( $h_{max}$ ) values of h-10 TIP and h-Tube surfaces with different micro/nano structures from the literature. The structures from the literature include microtube and TIP,<sup>[10]</sup> micropillar,<sup>[7a, 20]</sup> hierarchically 11 structured pillar (h-Pillar),<sup>[21]</sup> nanowire,<sup>[7d, 22]</sup> microcavity,<sup>[10, 23]</sup> microridge,<sup>[24]</sup> and 12 microchannel.<sup>[23, 25]</sup> All data shown in Figure 4 are summarized in Supporting Information 13 14 Table S1. Here we compared the data of SiO<sub>2</sub> surfaces, except nanowires based on silicon, to 15 compare the effects of structures excluding the effects of material properties. Our h-TIP 16 surfaces showed the highest simultaneous enhancement of HTC and CHF among all other

structures, as shown on the top-right corner of the plot. Yet, there are opportunities for further
 enhancement of h-TIP surfaces by optimizing microscale structure dimensions such as the
 outer diameter, pitch, and height of hierarchical tube and pillars, which were fixed in this
 work.

5





**Figure 4.** Regime map of CHF (q) rsub {CHF i) and maximum HTC  $(h_{max})$  values of different SiO<sub>2</sub> micro/nano structures and Si nanowires. This work with h-TIP shows the highest performance (upper right region) with simultaneous CHF and HTC enhancement.

10

In this work, we developed boiling surface structures that achieved superior pool boiling 11 12 performance via multi-scale control of vapor nucleation on hemi-wicking surfaces. Our 13 strategy included the minimization of bubble coalescence, promoting vapor nucleation and enhancing evaporation by engineering surfaces with separated tube clusters, microscale 14 15 cavities, and nanostructures, respectively. Meanwhile, capillary wicking performance was 16 maintained in the presence of dynamically interacting boiling bubbles. We conducted 17 saturated pool boiling experiments with water at atmospheric conditions and analyzed the 18 results, with supporting data from high-speed imaging of bubble dynamics. Our hierarchical 19 TIP surfaces (h-TIP) achieved significant HTC enhancement up to 389% as well as 138%

1 CHF enhancement compared to a smooth surface. This work provides surface design 2 guidelines for extreme pool boiling heat transfer, that is, the effective separation of nucleating 3 bubbles, enhanced evaporation by nanostructures, and exploiting capillary wicking are essential. We expect that our design guidelines can be adopted for industry-scale boiling 4 applications by creating surfaces using a scalable processes such as sandblasting;<sup>[26]</sup> for 5 6 example, a similar hierarchical structure can be created by sandblasting a surface using first a 7 larger abrasive and subsequently a smaller abrasive. Furthermore, physical insights obtained 8 in this work can be utilized in other applications such as electrochemical oxygen or hydrogen 9 evolution reaction, where surface-bubble interactions play a crucial role in their performance. 10 <sup>[27]</sup> The enhanced boiling performance promises significant energy savings in various boiling 11 applications, including steam power plants, desalination, thermal management of concentrated 12 photovoltaics, etc.

13

#### 14 Experimental Section

Surface fabrication: We first fabricated microscale structures through photolithography and 15 16 deep-reactive etching processes on silicon wafers that had 1 µm thermal oxide on both sides. A 2 µm layer of photoresist (SPR 700, Microposit) was spin coated with 3600 rpm for 30 17 seconds on one side of the wafers. After prebaking in a box furnace at 95 °C for 45 min, the 18 19 photoresist was exposed using an MLA150 Maskless Aligner. The resist was then developed in the developer (CD26, Microposit) followed by postbaking at 120 °C for 30 min. The 20 21 exposed silicon dioxide layer was etched by reactive ion etching (RIE) (Precision 5000, 22 Applied Materials) with CF<sub>4</sub> as a primary gas. Then, micropillars and microtubes were etched 23 in the bulk silicon with deep reactive ion etching (MESC Multiplex ICP, STS) based on SF<sub>6</sub> 24 and  $C_4F_8$ . The processed wafers were treated with oxygen plasma in the RIE chamber to 25 remove the C<sub>4</sub>F<sub>8</sub> passivation layers and organic contaminants. To create nanostructures over

1 the microfabricated surfaces, we sputtered a 500 nm copper (Cu) layer with a 50 nm titanium 2 (Ti) adhesion layer. The Cu layer was then oxidized in an alkali solution (NaClO<sub>2</sub>, NaOH, 3 Na<sub>3</sub>PO<sub>4</sub>, and deionized water with 3.75:5:10:100 wt.%) at 95 °C for 2 minutes, resulting in 4 sharp blade-like cupric oxide (CuO) nanostructures. Finally, the entire surfaces of hierarchical 5 structures were covered by a 20 nm silicon dioxide (SiO<sub>2</sub>) layer using atomic layer deposition.

6

7 Pool boiling characterization: The pool boiling setup consisted of a glass chamber with an 8 Ultern fixture at the bottom and a PEEK fixture at the top. The test sample  $(20 \times 20 \text{ mm}^2)$ 9 attached to an Ultem fixture with adhesive sealant (High-temperature RTV Silicone, 10 Permatex). The test sample was fabricated based on a 650-µm-thick silicon substrate. A rope 11 heater was used to maintain the saturation temperature in the glass chamber by preventing 12 heat loss. At the top PEEK fixture, an immersion heater and a reflux condenser were installed. 13 We first pre-heated de-ionized water in a separate electric kettle before introducing the water into the glass chamber. The immersion heater was used to degas the pre-heated water by 14 15 boiling for another 30 minutes before experimental measurements. During this degassing period, a heat flux of  $\approx 50$  W/cm<sup>-2</sup> was applied. The reflux condenser condensed water vapor 16 and allowed it to return back to the pool as liquid. A power supply (KLP 600-4-1200, Kepco) 17 provided power for the Joule heater of the  $10 \times 10 \text{ mm}^2$  heating area (A<sub>h</sub>) defined by the 18 19 serpentine Pt heater on the back side of the sample through Au/Pt electrodes. A multimeter 20  $(2110 5\frac{1}{2}, \text{Keithley})$  formed a serial connection with the power supply and the sample for the 21 current measurement across the heater (I), while the other multimeter (34401A, Agilent) iwas 22 connected in parallel for the voltage-drop measurement (V). The input heat flux was then evaluated as q } rsub {in} = (I \times V) / {A} rsub {h  $\dot{c}$ . The Pt backside heater also served as a 23 24 resistance temperature detector (RTD) for temperature characterization. Prior to pool boiling experiments, test samples were placed in a furnace with a high accuracy Pt RTD (Omega 25

Engineering, RTD-810) close to it. The heater resistance was then measured at six different 1 2 temperature set points. The dwell time at each set point was at least 2 hours to ensure thermal 3 equilibrium within the furnace. The resistance was measured with the four-wire method using a temperature input module (National Instruments, NI-9226 and cDAQ-9171). The 4 5 experimental data were fit with a linear model. The linear fit was used to convert the measured 6 resistance to the temperature of the heater during pool boiling experiments. Because of the 7 size mismatch between the actual heating area and the exposed surface area, there could be heat loss through the sample thickness. We numerically calibrated (COMSOL Multiphysics 8 9 5.3a LiveLink for MATLAB) the boiling heat flux and wall superheat based on the measured 10 input heat flux and the heater temperature. In the numerical simulation, we iterated the 11 calculations by changing the heat transfer coefficients at the boiling surface until the 12 calculation of the backside heater temperature matched the experimentally measured 13 temperature. All data points in the boiling curves were calibrated in this manner. Details of 14 the boiling test rig with schematics, experimental procedure, and measurement uncertainty are available in Section I of the Supporting Information. 15

16

#### 17 Supporting Information

18 Supporting Information is available from the Wiley Online Library or from the author.

19

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